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A COMPARISON OF TWO MELTING-PRESSURE EQUATIONS CONSTRAINED TO THE TRIPLE POINT USING DATA FOR ELEVEN GASES AND THREE METALS

ROBERT D. GOODWIN AND LLOYD A. WEBER

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IMPORTANT NOTICE

CONTENTS

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Robert D. Goodwin and Lloyd A. Weber

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Parameters have been determined by a least-squares method for the reduced Simon equation and for a new, empirical melting equation using data for H_2, D_2, T_2, Ne, Ar, Kr, Xe, N_2, O_2, CO_2, H_2O, Na, K, and Hg. The new equation, $(P-P_t)/(T-T_t) = A\exp(-\alpha/T) + BT$, represents experimental data with essentially the same accuracy as the Simon equation. It provides a sensitive difference method for graphical examination of data.

$\mathcal{A} \cup \tau \mathcal{H} \mathcal{O} \mathcal{R}$

1. Introduction

Experimental data for melting curves often have been represented by the original Simon equation [Simon and Glatzel, 1929; Simon, Ruhemann, and Edwards, 1930],

$$P = a + bT^c \tag{1-a}$$

with three parameters, omitting the triple-point datum. Since a number of triple-point determinations now are available, it is appropriate to establish the two parameters of the reduced Simon equation [Simon, 1937; 1953],

$$P - P_t = P_o \left[(T/T_t)^c - 1 \right] \tag{1}$$

as constrained to the triple point (subscript t), using a uniform computational method based on relative, rather than absolute deviations for all substances.

Whereas (1) has been derived with certain assumptions from modern theory of solids [Gilvarry, 1956; Glass, 1963; Salter, 1954; Voronel, 1948, 1959], real or apparent difficulties are encountered in the accurate empirical representation of some data, e.g. [Michels and Prins, 1962; Pistorius, Pistorius, Blakey, and Admiraal, 1963], and for some metals the theoretically derived relation

$$c = (6\gamma + 1)/(6\gamma - 2)$$

(γ = Grueisen's constant) is not confirmed experimentally [Strong and Bundy, 1959].

In view of these difficulties, we have at the same time examined further the empirical equation recently used for interpolation near the triple point of hydrogen [Goodwin, 1962; Goodwin and Roder, 1963],

$$y = A \exp(-a/T) + BT, \tag{2}$$

where

$$y \equiv (P - P_t)/(T - T_t).$$

For this examination, substances other than low-boiling gases are included for which, however, the triple-points may not be accurately known. It is possible to deduce that (2) implies a temperature-dependence of the Grueisen constants, not found in the assumptions of the latter equation of state. Gilvarry, however, has concluded that a small temperature-dependence of these constants may be expected [Gilvarry, 1956]. Figure 1 illustrates behavior of the experimental function y with data for H_2, D_2, N_2, Ar, and CO_2. The small temperature-dependence of this function (as compared with the pressure

P) provides a direct method for graphical examination of the relative deviations or precision of a set of data, prior to the tedious iterative determination of parameters either for (1) or (2).

Concerning helium, for which no triple-point exists, we merely note, for example, that for $-(a/T) \ll 0$ at very low temperatures, (2) may be reduced to

$$P = P_t - (BT_t) \cdot T + BT^2 \qquad (2\text{-}a)$$

and that this form is the same as used by Mills, Grilly and Sydoriak (1961) near the melting pressure minimum for He^3 in the range $0.3 \leq T \leq 0.5°K$, namely,

$$P = 32.42 - 21.25 T + 32.20 T^2.$$

Equation (1), on the other hand, clearly is monotonic.

2. Data and Computational Method

For each substance, the self-consistent data of but one investigator have been selected for present purposes. Original, unpublished data on hydrogen, deuterium, tritium, neon, nitrogen, and oxygen were generously given to us by Edward R. Grilly [Mills and Grilly, 1955; Mills and Grilly, 1956]. Argon, on the other hand, has been selected for illustration using data of three investigators over a wide range of pressures [Bridgman, 1935; Lahr, and Eversole, 1962; and Michels and Prins, 1962]. Further references to experimental data are given by authors cited here. Triple-point constants used for the present computations are given in table 1. All pressures are in atmospheres [Hilsenrath, 1955]. Absolute temperatures below 80°K are on the NBS 1955 low-temperature scale [Goodwin and Roder, 1963].

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Since relative, rather than absolute, error is roughly constant in the experimental determination of melting pressures, the least-squares method (with trial variation of one parameter) has been applied to constant or nearly constant forms of the above equations,

$$(P - P_t) / \left[(T/T_t)^c - 1 \right] = P_o, \qquad (1\text{-}b)$$

$$(P - P_t)/(T - T_t) \, T = A T^{-1} e^{-a/T} + B. \qquad (2\text{-}b)$$

In (2-b), all three constants have been treated as adjustable parameters, in contrast to the earlier treatment for hydrogen isotopes [Goodwin, 1962].

Computational results include the root mean square (r. m. s.) absolute deviation in atm, and the r. m. s. relative deviation in percent, defined respectively by

$$D \equiv \left[\sum (\Delta P)^2 / n \right]^{1/2}, \qquad (3\text{-}a)$$

$$\Delta \equiv \left[\sum \delta^2 / n \right]^{1/2}, \qquad (3\text{-}b)$$

where

$$\delta \equiv 100 \, (P_{calc} - P)/P \qquad (3\text{-}c)$$

for a number $\underline{n}$ of datum points, and where ΔP is the difference between calculated and experimental values. The form of (3-a) is the same as used by Mills and Grilly [1956]. Minima in D and Δ, however, generally do not occur for the same set of parameters. Those corresponding to the minimum relative deviation, Δ, are given in the following results, in contrast to those for a minimum in D given by Mills and Grilly [1956]. To examine detailed behavior

near the triple point, the average of absolute values of deviations from the first six data of each investigator at the lowest pressures are given in percent in table 2 under the symbol $\bar{\delta}$.

3. Results

Table 2 presents the sources of data and the number of points, n, used in the least-squares determination of parameters, and the maximum pressure of these data. There follow the parameters and deviations for each equation. For hydrogen on lines 1 and 2, two different sets of data are compared. Argon is examined with various data. Line 6 is for the mutually consistent data of Michels and Prins [1962] and of Bridgman [1935]. On line 7 (2) has been fitted to synthetic data computed by (1), line 6. Line 8 includes, also, the data of Lahr and Eversole to 18,000 atm [Lahr and Eversole, 1962], whereas line 9 employs the latter data only.

A comparison of relative deviations, Δ, in table 2 shows in general that (2) represents the data as well as, or, for some substances, somewhat better than (1). The improvement is clear for the hydrogen data on line 1, for argon on line 6, for xenon, and for water.

Equation (2) contains a term which may be compared with the internal pressure, P_o, of (1) at $T = T_t$. The ratio of these terms for the gases through O_2 in table 2 is found to be

$$(AT_t/P_o) \exp (-a/T_t) = 1.2 \pm 0.1$$

with the exception of xenon for which the ratio is 1.70.

For xenon, in table 3, the comparison of equations is given at all points, since Michels and Prins found it necessary to fit the data by (1-a), independently, in two regions [Michels and Prins, 1962].

Whereas (2) gives a better average representation than (1), it does not overcome the systematic deviation seen in this table.

Some of the results for argon are presented graphically. The ordinate of the figure 2 logarithmic plot was computed with the internal pressure of Michels and Prins for (1-a), while the slope of the extrapolated straight line corresponds to their values of $\underline{c}$ [Michels and Prins, 1962]. Included for comparison are data of Bridgman [1935], of Lahr and Eversole [1962], and of Robinson [1954]. As compared to the pressure at 360°K (near 18,000 atm.), given by (1), line 6 of table 2, the smoothed function of Lahr and Eversole yields a value 7.0% lower, and (2), line 6 of table 2, yields a value 3.5% higher, the latter equation being shown by the dashed curve of figure 1.

Figure 3 for argon is derived from (2), using parameters from line 6 of table 2. The straight line represents (2). The dashed line represents (1). Datum points are the same as on figure 2. Figure 4 is similar, except that all constants are from line 8 of table 2. This type of difference plot provides a highly sensitive method for graphical examination of the precision of experimental data.

Table 4 for argon presents relative deviations of both equations from the data of Bridgman [1935]; Lahr and Eversole [1962]; and Michels and Prins [1962], line 8 of table 2 to 18,000 atmospheres. It is seen that the two equations give comparable representations of these combined data to 18,000 atm.

Table 5 for argon compares first derivatives, $s \equiv dP/dT$, of the two equations in terms of percent difference, $100(s_2 - s_1)/s_1$, in which subscripts refer to the equations. Under heading (a) are given results for the consistent data of line 6 of table 2, whereas, under (b), the results correspond to the apparently less consistent data of line 8

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of table 2. For each case, it is seen that the first derivatives of the two equations are comparable over the range of the data.

Table 6 presents a comparison of the Simon constants recently determined by S. E. Babb, Jr. [1963], by a method of absolute deviations, with those determined in the present report by a method of relative deviations.

Since importance has been attached to the fact that (1) is a "reduced" form [Simon, 1937], the reduced constants for (2) are examined in table 7. The following ranges of values are found for the gases H_2 through N_2 of table 2,

$$\frac{a/T_t}{0.04 \to 0.4} \qquad \frac{AT_t P_t}{(3 \to 17) \cdot 10^3} \qquad \frac{BT_t/P_t}{(0.1 \to 1.3) \cdot 10^2} .$$

4. Summary and Conclusions

The two parameters of the reduced Simon equation have been determined for a number of gases and some metals by a uniform method which assumes that relative rather than absolute uncertainties are roughly constant in the experimental pressures.

Some current empirical and theoretical difficulties with the Simon equation, already mentioned, have led to examination of a new, empirical equation with three parameters. The latter equation is presented as a result of the remarkable equivalence to the Simon equation found here in the empirical representation of data, even though no simple mathematical equivalence between the equations is apparent.

It may be concluded that (1) will be preferred to (2) for its simplicity and qualitative theoretical support, except where the latter gives improved empirical representation of data, or until such time as it receives some theoretical interpretation. Equation (2), however, provides a sensitive difference method for graphical examination of the precision of data.

5. Acknowledgements

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Table 1. Triple-point constants used.

Substance	T_t, °K	P_t, atm.	Reference
$n\text{-}H_2$	13.947	0.071	[31]
$n\text{-}D_2$	18.72	0.169	[10]
$n\text{-}T_2$	20.61	0.213	[10]
Ne	24.544	0.427	[11]
Ar	83.812	0.685	[17, 18]
Kr	115.745	0.724	[18]
Xe	161.364	0.806	[18]
N_2	63.146	0.124	[12]
O_2	54.353	0.0015	[13]
CO_2	216.577	5.11	[17, 12]
H_2O	354.75*	21,260.*	[22]
Na	370.75†	0.9684†	[3]
K	335.65†	0.9684†	[3]
Hg	234.32†	0.0	[17]

* Triple-point ice VI + ice VII + liquid.

† The melting-point of original data.

Table 2. Constants and deviations for the two equations

Line	Sub-stance	Reference	n	P_{max}	c	P_o	D	$\bar{\delta}$	Δ	α	A	B	D	$\bar{\delta}$	Δ
	Data				Equation 1					Equation 2					
1	$n\text{-}H_2$	[31]	15	608	1.930	216.72	2.7	0.52	0.75	6.2	32.859	.60768	0.8	0.50	0.43
2	$n\text{-}H_2$	[20]	26	3,600	1.795	244.89	15.7	2.06	1.41	4.1	28.803	.67444	10.4	2.43	1.45
3	$n\text{-}D_2$	[20]	23	3,500	1.815	406.07	7.7	0.56	0.49	4.4	33.055	.67890	4.6	0.65	0.44
4	$n\text{-}T_2$	[20]	15	3,000	1.780	509.88	3.6	1.32	1.14	1.0	30.869	.72807	6.5	1.34	1.16
5	Ne	[19]	36	3,400	1.630	985.06	6.2	0.47	0.46	4.0	59.334	.59879	5.7	0.46	0.47
6.	Ar	[4,18]	16	5,800	1.594	2,087.7	3.3	0.03	0.09	7.2	33.275	.10971	1.0	0.03	0.05
7.	Ar	- -	16	5,800	- - - -	- - - -	- - -	- - -	- - -	9.0	34.294	.10596	1.32	0.03	0.04
8.	Ar	[4,14,18]	30	18,000	1.521	2,213.3	89.9	0.75	1.47	24.0	45.413	.06264	75.0	0.39	1.33
9	Ar	[14]	14	18,000	1.510	2,242.2	108.1	1.95	1.80	48.0	56.334	.04234	119.2	1.66	1.57
10	Kr	[18]	14	1,500	1.615	2,349.0	0.1	0.12	0.11	5.0	24.949	.07680	0.2	0.12	0.11
11	Xe	[18]	19	1,500	1.710	2,352.3	2.1	0.46	0.42	71.0	38.580	0.0	1.0	0.29	0.26
12	N_2	[19]	24	3,500	1.790	1,586.2	2.1	0.30	0.22	7.0	33.133	.24087	2.0	0.31	0.21
13	O_2	[19]	16	3,500	1.755	2,666.8	5.1	0.60	0.47	14.0	83.374	.38743	5.4	0.59	0.44
14	CO_2	[15]	25	2,800	3.1	3,178.3	5.4	2.48	1.65	0.0	- 7.05304	.242083	4.9	2.55	1.64
15	H_2O	[22]	26	196,000	4.8	6,048.3	4370.0	2.30	5.13	450.	-5546.02	4.80652	3005.	2.30	3.58
16	Na	[3]	12	11,600	3.6	11,409.0	22.1	0.76	0.74	380.	-1530.54	1.78578	26.2	0.74	0.62
17	K	[3]	12	11,600	4.4	4,224.4	31.3	0.37	0.44	280.	- 503.249	.815969	39.4	0.35	0.49
18	Hg	[16]	13	3,000	0.76	59,249.	3.6	0.99	0.94	260.	-8872.22	13.3083	2.4	0.94	0.88

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Table 3. Comparison of (1) and (2) for Xenon

T	P	δ_1	δ_2
161.554	5.53	0.33	0.00
162.439	27.49	0.66	0.39
163.375	50.82	0.67	0.45
164.171	70.95	0.37	0.19
167.154	147.31	-0.23	-0.28
171.455	259.23	-0.51	-0.42
176.978	405.89	-0.64	-0.42
184.004	596.25	-0.56	-0.26
191.144	794.08	-0.39	-0.09
197.868	984.49	-0.22	0.02
203.000	1132.20	-0.06	0.09
207.205	1255.18	0.05	0.10
211.142	1371.51	0.17	0.11
215.264	1494.53	0.30	0.11
		0.37	0.21

Table 4. Comparison of eqns. (1) and (2) for Argon.*

T	P	δ_1	δ_2
Michels and Prins			
85.532	69.39	1.07	-.71
87.458	147.46	.89	-.65
89.110	215.06	.88	-.47
93.543	400.40	.69	-.20
98.129	597.46	.54	.06
102.587	794.12	.41	.25
106.796	984.49	.28	.37
109.921	1129.13	.14	.39
112.526	1251.52	.03	.41
115.095	1373.85	-.06	.41
117.649	1497.02	-.16	.41
Bridgman			
106.4	967.8	.14	.21
144.9	2903.5	.92	.08
161.9	3871.4	-1.52	-.55
177.8	4839.2	-2.15	-1.29
192.9	5807.0	-2.67	-1.96

T	P	δ_1	δ_2
Lahr and Eversole			
137	2389	3.01	3.99
182	4972	.29	1.13
188	5417	-1.23	-.46
197	6249	-5.46	-4.82
226	7690	1.35	1.69
245	9100	.02	.13
262	10250	.65	.57
278	11366	1.17	.93
284	11898	.45	.16
288	12236	.16	-.17
291	12440	.37	.01
299	12960	1.12	.69
317	14413	.81	.25
360	18040	.35	-.43
		0.97	0.80

* Constants from line 8 of table 2.

Table 5. Comparisons of $s \equiv dP/dT$ for Argon

T	$100\,(s_2 - s_1)/s_1$	
	(a)	(b)
83.812	+.06	-2.02
83.900	+.07	-2.00
84.000	+.07	-1.97
84.500	+.06	-1.83
85.000	+.06	-1.70
86	+.06	-1.44
87	+.06	-1.21
88	+.05	- .98
90	+.04	- .58
100	-.06	+ .80
110	-.15	+1.42
120	-.21	+1.61
140	-.13	+1.34
160	+.18	+0.74
180	+.69	+0.10
200	+1.35	-0.47
240	-----	-1.30
280	-----	-1.71
320	-----	-1.76
360	-----	-1.54

(a) Constants from line 6 of table 2.

(b) Constants from line 8 of table 2.

Table 6. Comparison of Simon Constants of S. E. Babb, Jr. [1963] with those of table 2.

Substance	$(P_o)_{Babb}/(P_o)_{table\ 2}$	$(c)_{Babb}/(c)_{table\ 2}$
$n\text{-}H_2$	1.1051	0.9716
$n\text{-}D_2$	1.0443	0.9847
$n\text{-}T_2$	1.0258	0.9911
Ne	1.0395	0.9815
Av	0.9992	0.9994
Kr	0.9983	1.0012
Xe	1.0951	0.9294
N_2	0.9996	1.0006
O_2	1.0114	0.9929
CO_2*	1.2421	0.8387
H_2O*	2.1180	0.6479
Na	1.0355	0.9814
K	0.9975	1.0091
Hg*	0.6366	1.5487

* Different data used by respective authors.

Table 7. Approximate reduced constants for equation (2)

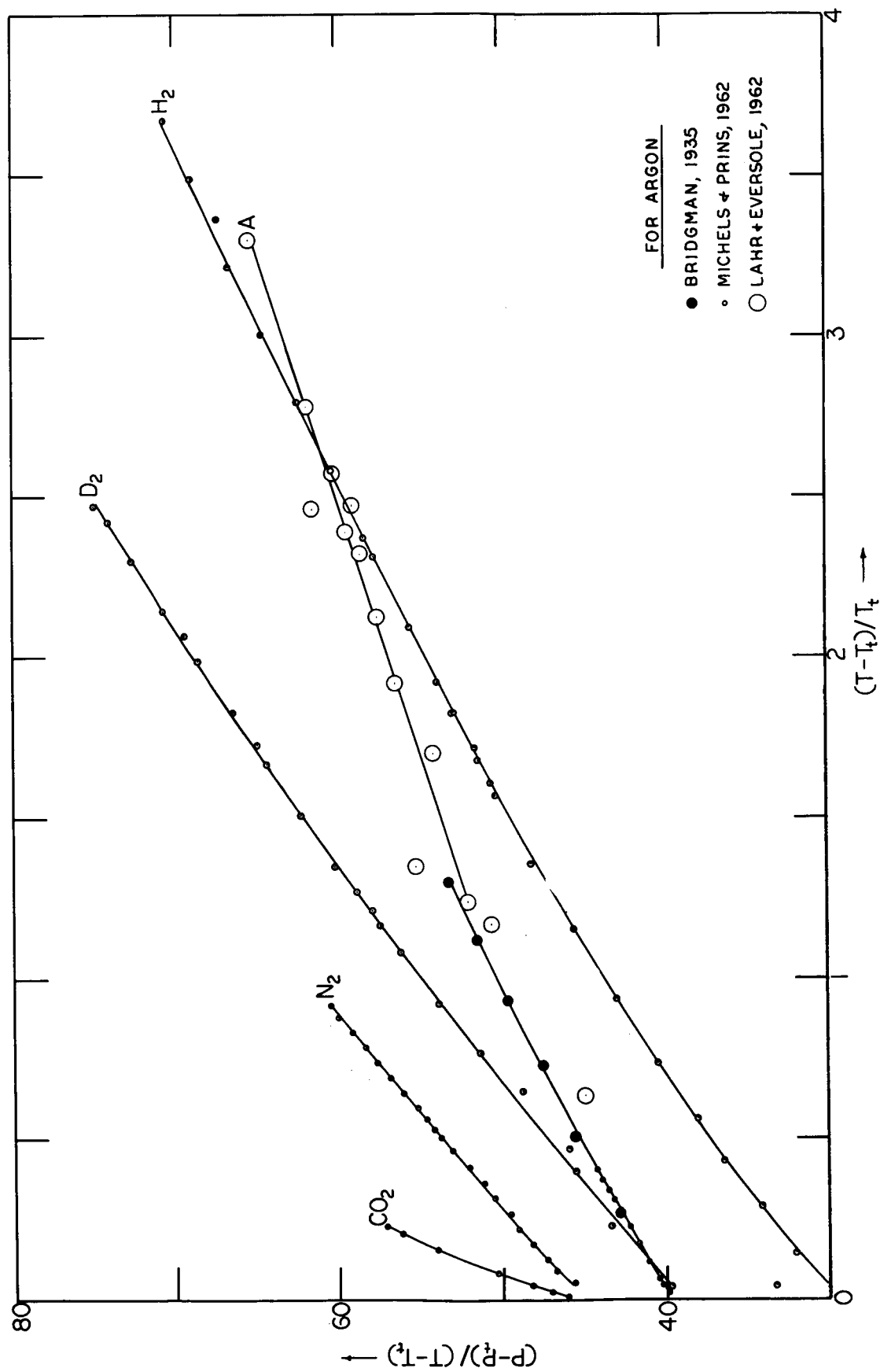| Line*	Substance	a/T_t	$(AT_t/P_t) \cdot 10^{-3}$	$(BT_t/P_t) \cdot 10^{-2}$
1	H_2	0.445	6.45	1.194
2	H_2	0.294	5.66	1.325
3	D_2	0.235	3.66	0.752
4	T_2	0.049	2.99	0.704
5	Ne	0.163	3.41	0.344
6	Ar	0.086	4.07	0.134
7	Ar	0.107	4.20	0.130
8	Ar	0.286	5.56	0.076
9	Ar	0.573	6.89	0.052
10	Kr	0.043	3.89	0.120
11	Xe	0.440	7.72	0
12	N_2	0.111	16.87	1.227
13	O_2	0.258	3021.	140.
14	CO_2	0	-0.30	0.103
15	H_2O	1.27	-0.09	.0008
16	Na	1.03	-586.	6.84
17	K	0.83	-175.	2.83

* Lines correspond to table 2.

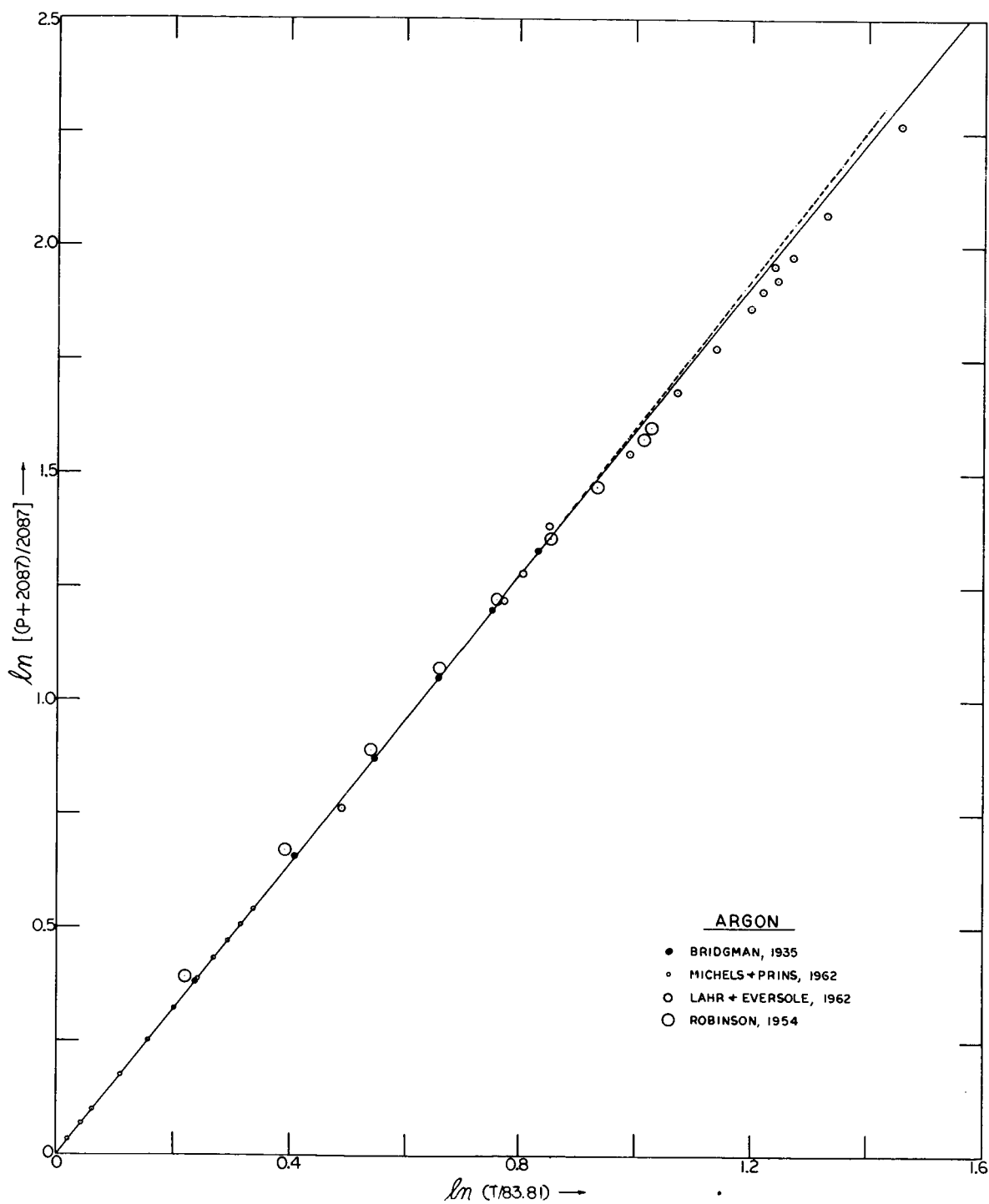Figure 1.   The function $(P-P_t)/(T-T_t)$ vs. $(T-T_t)/T_t$.

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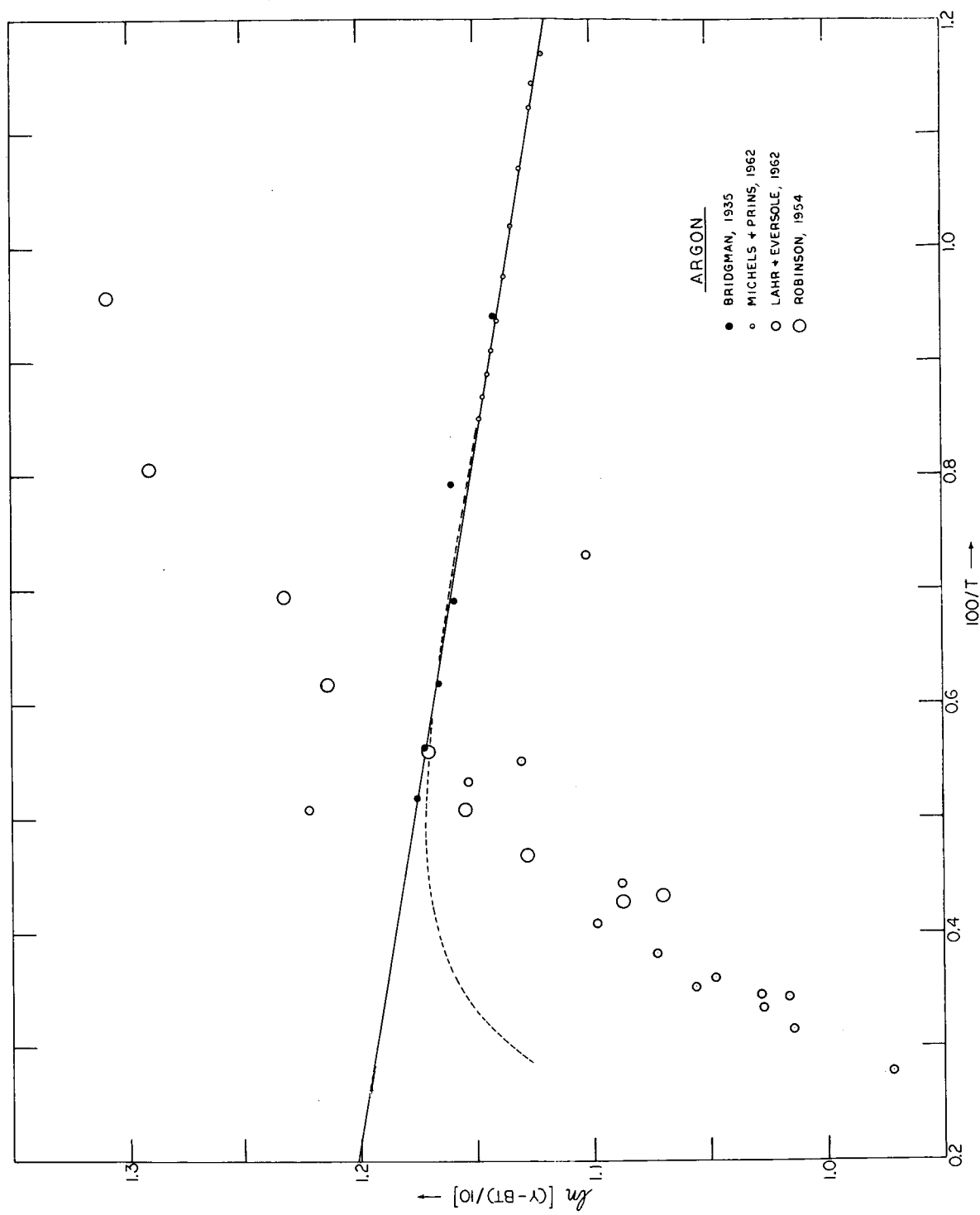Figure 2.   Logarithmic plot of equation (1) for argon using the value P_o = 2087 atm.

Figure 3.   Logarithmic plot of equation (2) for argon
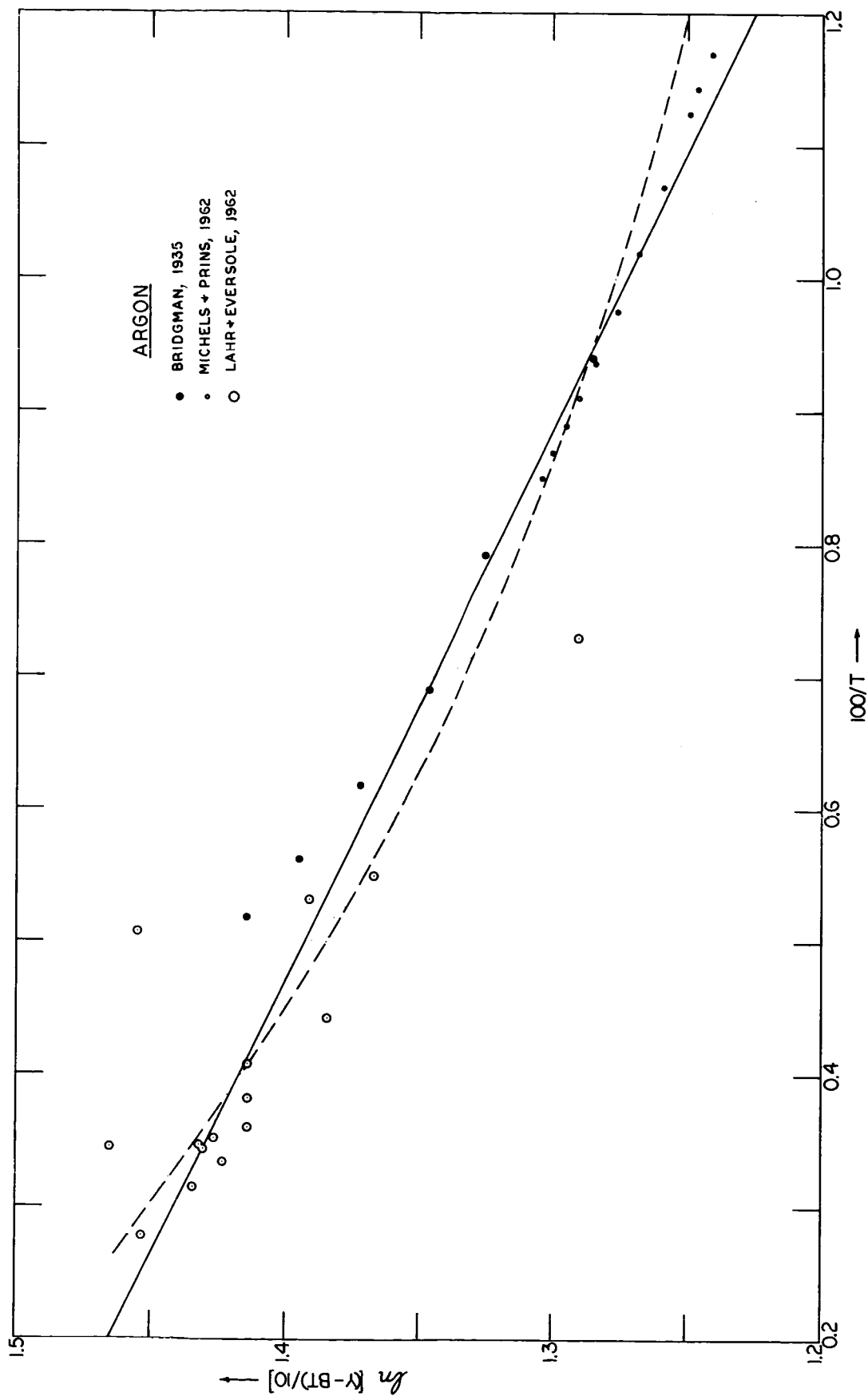using the value   B = 0.10971 atm deg^{-2}

Figure 4.   Logarithmic plot of equation (2) for argon
using the value B = 0.06264 atm deg^{-2}.

ARGON

● BRIDGMAN, 1935

○ MICHELS + PRINS, 1962

○ LAHR + EVERSOLE, 1962

$100/T$ →

← $\ln[(\chi-BT)/10]$

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